

# Bulk elastic fingering instability in Hele-Shaw cells

B. Saintyves,<sup>1,\*</sup> O. Dauchot,<sup>2</sup> and E. Bouchaud<sup>1,2</sup>

<sup>1</sup>*CEA-Saclay, IRAMIS, SPEC, F-91191 Gif-sur-Yvette Cedex, France*

<sup>2</sup>*ESPCI-Paris Tech, PSL\*, UMR Gulliver, EC2M,  
10 rue Vauquelin, 75231 Paris Cedex 05, France*

(Dated: July 3, 2012)

We demonstrate experimentally the existence of a purely elastic fingering instability which arises when air penetrates into an elastomer confined in a Hele-Shaw cell. Fingers appear sequentially and propagate within the bulk of the material as soon as a critical strain, independent of the elastic modulus, is exceeded. Their width depends non-linearly on the distance between the confining glass plates. A key element in the driving force of the instability is the adhesion of layers of gels to the plates, which results in a considerable expense of elastic energy during the growth of the air bubble.

PACS numbers: 61.41.+e, 62.20.-x, 68.35.Np, 68.35.Gy

Bulk fingering instabilities in viscous liquids confined in Hele-Shaw cells, commonly known as the Saffman-Taylor instability, have given rise to considerable experimental [1] and theoretical effort [2]. In the context of the liquid to solid transition (gels [3], foams [4], yield stress fluids [5–7] and Maxwell liquids [8]), this instability translates into a fingering to fracture transition.

Purely elastic instabilities in a confined geometry have received much less attention. An elastic fingering instability has been observed during the peeling of a thin layer of elastomer from a rigid substrate [9, 10] or in probe tack experiments [11] where a semi-spherical probe in contact with the soft solid is pulled up at a constant speed. In most cases, the instability settles on the crack line, which is observed to progress at the interface between the elastomer and the rigid body it separates from [9, 12]. The energetic balance involving adhesion, elastic energy stored within the film and rigidity of the plate favors a meandering instability of the crack front, which has been fully characterized experimentally and theoretically [13, 14]. In contrast, Shull et al. [15, 16] have observed fingers propagating within the bulk of an acrylic tri-block copolymer gel. They demonstrated the reversibility and the non hysteretic character of the process. However, to our knowledge, there has been no quantitative characterization of the observed patterns yet, and one dramatically lacks experimental data to constrain the possible mechanisms at the origin of this instability.

In this letter, we demonstrate the existence of a large scale – centimetric – elastic fingering instability arising within the bulk of a polyacrylamide gel confined in a Hele-Shaw cell. The instability appears when a critical strain, independent of the shear modulus is exceeded. Fingers appear sequentially, and, ultimately, lead to the spectacular flower-shaped pattern displayed in Fig. 1. The size of the fingers increases non-linearly with the cell thickness, while the number of fingers decreases sub-linearly. The latter is independent of the shear modulus.

**Materials and methods.** We use polyacrylamide gels made from acrylamide monomers and bis-acrylamide

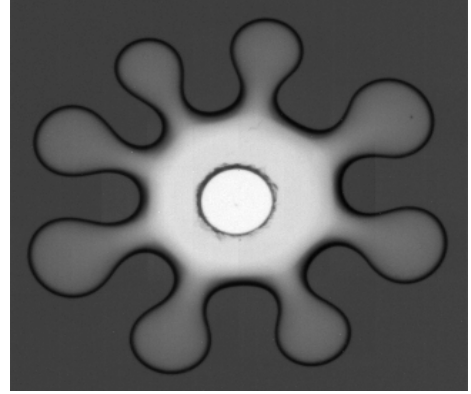


FIG. 1: **Elastic fingering instability.** Pattern of the destabilized interface ( $G = 650$  Pa;  $b = 2.1$  mm;  $Q = 126$  ml/min)

cross-linkers [17]. The relative concentrations of these two components control the shear modulus of the gel from  $G = 80$  to  $1060$  Pa. The strain to fracture of the produced materials decreases with  $G$ , but exceeds 300% in all cases. Adding dye to the gel allows to see where the material sticks to the glass plates. Cyclic shear rheology of the gels with dye indicates a very small ratio of the elastic modulus to the loss one:  $G''/G' < 10^{-3}$  for frequencies ranging from  $0.01$  Hz to  $100$  Hz.

Two different experimental setups are used. Setup 1 (Fig. 2 left-(a)) is a classical Hele-Shaw: it consists in two  $10$  mm thick glass plates of lateral sizes ( $250$  mm  $\times$   $250$  mm) separated by thin spacers of thickness  $b \in [0.5 - 5]$  mm. Setup 2 (Fig. 2 left-(b)) is an original design, which consists in a sealed Hele-Shaw cell made of two  $10$  mm thick glass plates of lateral sizes ( $250$  mm  $\times$   $125$  mm), with two opposite mobile walls, acting as pistons, and pulled at a prescribed velocity  $V$  by synchronized step motors. The gap is fixed to the value  $b = 2.1$  mm. This setup is closer to classical tensile tests in solid mechanics. In both experiments the cells are filled with polyacrylamide before gelation. During this process, we maintain an initial air bubble of controlled diameter ( $D_0 = 23$  mm in Setup 1,  $D_0 = 15$  mm in Setup 2). In Setup 1, air is blown with

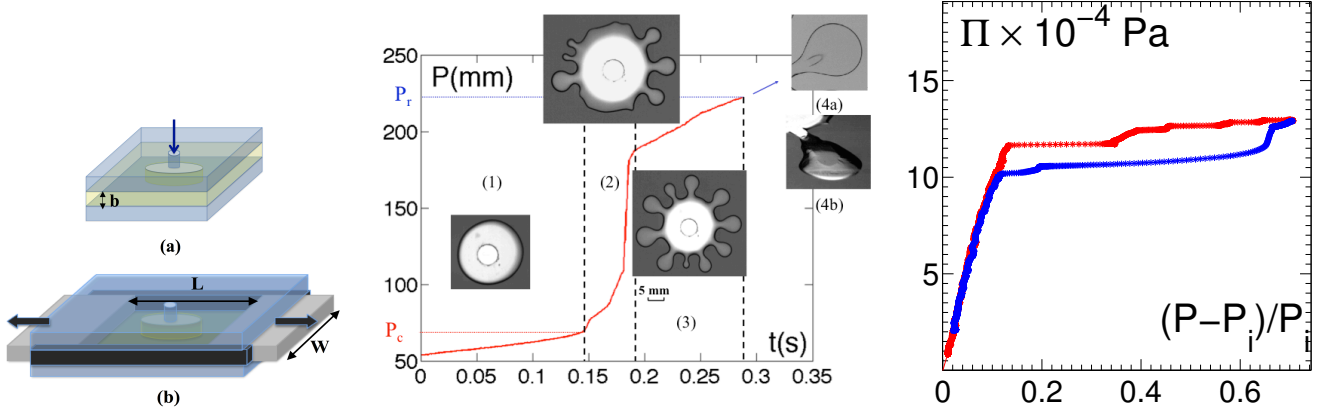


FIG. 2: **Setups and basic observations:** (color online) **(Left):** (a): Setup 1, classical Hele-Shaw cell with air injection at the center. (b): Setup 2, sealed Hele-Shaw cell with two moving walls acting as pistons. **(Center):** Sketch of the instability observed in Setup 2 ( $G = 500$  Pa,  $b = 2.1$  mm): Evolution of the perimeter  $P$  of the pattern as a function of time, exhibiting four distinct stages: (1) Circular growth; (2) Nucleation of the fingers; (3) Swelling of the fingers; (4a) Crack appearing in the gel layer leading to (4b) an interfacial fracture.  $P_c$  (respectively  $P_r$ ) is the bubble perimeter at the onset of fingering (reps. just before fracture). **(Right):** Reversibility and hysteresis observed in setup 1 ( $G = 840$  Pa,  $b = 1.4$  mm): Pressure  $\Pi$  vs. strain  $\gamma = (P - P_i)/P_i$ , (red curve : inflation; blue curve: deflation).

a syringe pump in order to grow this bubble. The pressure varies from 0 to 1.5 bar. In Setup 2, the depression induced by the motion of the pistons sucks air into the gel. Because polyacrylamide is incompressible, the speed and size of the pistons impose an air flow rate equal to:  $Q = 2WbV = 126$  ml/min. In both cases, the loading rates are such that the material can safely be considered as purely elastic.

Most observations are top views of the cell recorded at 50 Hz for Setup 1 and 1500 Hz for Setup 2, which gives an idea of the robustness of the instability over a wide range of interface speeds. We measure the thickness of the gel layers through light absorption. When performing such measurements, we use a silicon oil (Rhodorsil V20), which is immiscible with the gel but has a similar refractive index and therefore avoids refraction. The observed patterns show no differences with the one obtained when injecting air: The interfacial tension between the injected fluid and the gel is not a relevant parameter.

*Experimental results* The same scenario holds in both setups and can be summarized as follows (Fig. 2-center and Supp. Materials [24]). The evolution of the perimeter  $P$  of the pattern allows to distinguish four stages. During stage (1), a circular bubble grows. Shades of gray behind the dark line of the front show the existence of layers of gel adhering to the glass plates. In stage (2), fingers burst out successively, with an experiment-dependent order of appearance, and with a speed much larger than the velocity of circular extension. This leads to a sharp increase of the perimeter. Stage (3) corresponds to the swelling of the fingers once they are all formed, and the increase of the perimeter is much slower again. Then, in stage (4) the layer of gel behind the tip

breaks, leading to an interfacial crack.

Prior to the interfacial fracture stage, the phenomenon is completely reversible: Fingers deflate when the tensile stress is released, and the initial bubble is recovered. The reversibility of the process clearly demonstrates the purely – possibly non linear – elastic nature of the instability. Figure 2 (right) displays the pressure  $\Pi$  as a function of strain  $\gamma = (P - P_i)/P_i$ , where  $P_i = \pi D_0$  is the initial bubble perimeter. It shows that the phenomenon is slightly hysteretic. When air is injected a second time, the pressure follows the red curve again, which discards the hypothesis that the material has undergone damage during the first cycle.

By filming the cell at an angle, we could see a clear meniscus on all fingers. Besides, as already shown in Fig. 2(center), interfacial cracking occurs at a late stage, after the full development of the instability. There two distinct fronts are easily observable, one corresponding to the interfacial crack, the other one to a deformation of the bulk ahead of it (Fig. 2(center) image 4(b)). Further quantitative evidence of this 3D character is provided in Fig. 3, where color encodes the gel thickness measured from light absorption (Fig. 3a-b). Before a finger bursts out, the interface is already locally deformed. The gel thickness increases with the distance to the injection point. The finger first develops with a constant width and almost with a constant thickness as further illustrated in Fig. 3(c), where the finger profiles within the thickness of the cell have been plotted at constant time intervals, assuming a symmetric shape. The finger grows up to a certain size where only the extremity is swelling while the back, forming a saddle, does not evolve any more. This early stage is followed by a regime where both the

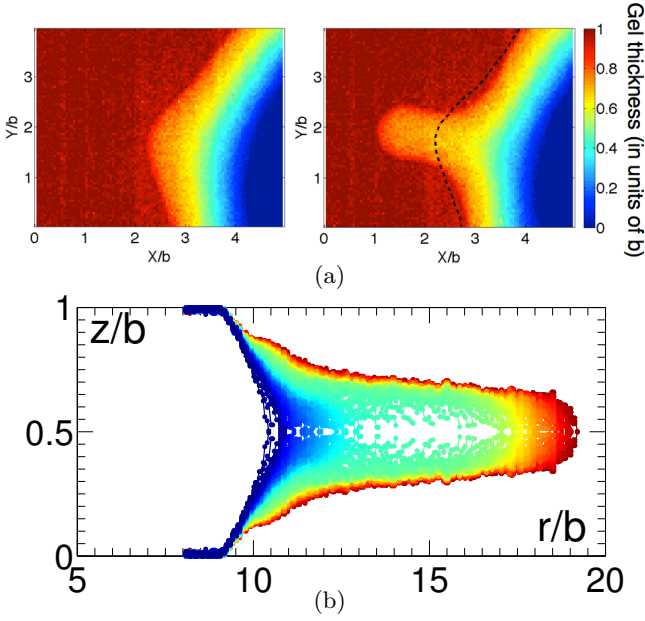


FIG. 3: **Formation of a finger:** (color online) **(a):** Top view before (left) and after (right) the finger has formed. The initial front (black dotted line) recedes in the finger vicinity. Color bar: thickness of the gel in  $b$  units. **(b):** Thickness profiles of a finger developing in time. Color codes 137 time steps from blue to red separated by 0.6 ms.  $G = 100$  Pa.

extremities and the “saddle” parts of the fingers increase in size. Note that the formation of the finger is accompanied by a relaxation of the elastic strain in its vicinity, where the front is seen to recede (Fig. 3(a-b)). The two phenomena arise instantaneously - within experimental accuracy -, and are far more rapid than the growth rate of the whole pattern. In some cases, one observes small fingers, which start to develop and which eventually recede, if they are surrounded by larger ones (see inset(3) of fig. 2(center)). Again this is a clear signature of the elastic nature of the process.

As can be seen from the perimeter temporal evolution (Fig. 2-center), the instability sets in when the strain exceeds a critical value  $\gamma_c$  which depends on the perimeter of the initial bubble  $P_i$  as  $1/P_i$ . For a given  $P_i$ ,  $\gamma_c$  does not depend on the shear modulus (Fig. 4(a)), in contrast with the strain to fracture  $\gamma_r = (P_r - P_i)/P_i$ , which decreases with  $G$ . This latter behavior tends to indicate that the elongational stress to failure tends to decrease with  $G$ , which is usually the case for solid brittle materials. The characteristic wavenumber of the pattern, as computed from the power spectrum of the interface  $r(\theta)$  before fracture ( $r$  and  $\theta$  are the polar coordinates of the points lying on the pattern border) does not depend on  $G$  either (Fig. 4(b)).

Finally experiments performed with Setup 1, for which the gap  $b$  can be varied easily, allow to estimate the role of confinement. The width  $\delta$  of the fingers at their root, measured when they are just formed (Fig. 5(a)), and the

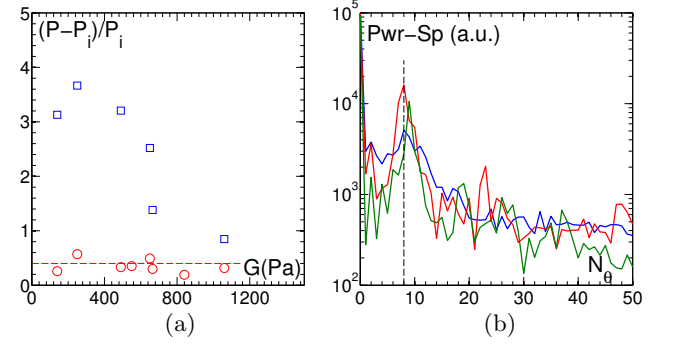


FIG. 4: **Dependence on the shear modulus** (color online) **(a):** Critical strain at the onset of fingering,  $\gamma_c$  ( $\circ$ ), and strain to fracture  $\gamma_r$  ( $\square$ ), as a function of the shear modulus  $G$  (setup 2). **(b):** Power spectrum of  $r(\theta, t)$ , averaged over time during stage (3) for three different bulk moduli (blue):  $G = 140$  Pa, (red):  $G = 660$  Pa, (green):  $G = 1060$  Pa (setup 2).

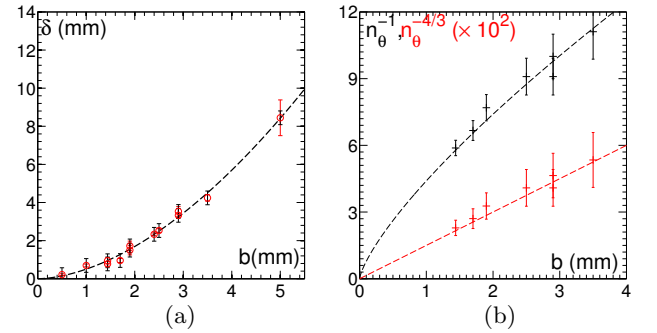


FIG. 5: **Influence of the gap** (color online) **(a):** Width of the fingers at their root,  $\delta$  (main panel). **(b):** Asymptotic inverse wave number  $n_\theta^{-1}$  (+), and  $n_\theta^{-4/3}$  (++) vs.  $b$ : the continuous lines are indicative fits of the data, by respectively a power law  $b^{7/4}$  (main panel),  $b^{3/4}$  and a linear fit.

inverse wavenumber of the instability  $n_\theta^{-1}$  (Fig. 5(b)) exhibit nonlinear dependence on  $b$ . Indicative fits suggest power law-like behaviors  $\delta \sim b^{7/4}$  and  $n_\theta^{-1} \sim b^{3/4}$ . Fig. 6(a) displays a spatio-temporal diagram of the local strain  $\gamma_{loc} = (r(\theta, t) - r(\theta, 0))/r(\theta, 0)$  recorded in Setup 1 for a gap of 2.9 mm. Quite remarkably, despite their sequential appearance, fingers end up regularly spaced, without having drifted. One can even identify the location of a missing finger ( $\theta \simeq -2\pi/3$  in Fig. 6(a)), which would eventually develop, if the pattern were not to fracture before. This suggests the existence of a well defined wavenumber. Fig. 6(b) shows the evolution of strain both along each finger, and between the fingers. The strain is evenly distributed until the fingers start to grow (between  $t \simeq 18$  s and  $t \simeq 25$  s). Then blue curves take off, showing stress concentration at each finger tip, while red curves decrease, showing that this local stress concentration is accompanied by a stress release in between fingers. This release is what causes the front to recede in the close vicinity of fingers, as shown above (Fig. 3).

*Discussion* We have reported experimental evidence of a bulk fingering instability arising in a confined layer of a

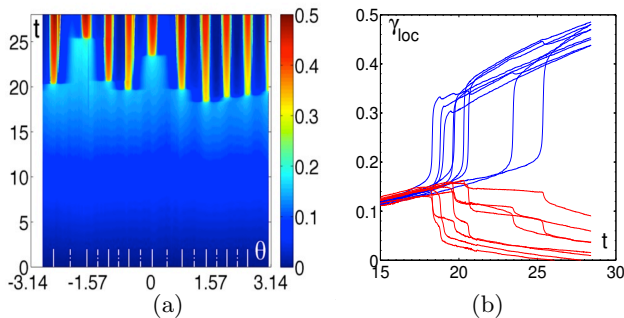


FIG. 6: **Spatio-temporal diagram** : (color online) (a): Local strain  $\gamma_{loc}(\theta, t)$ ; (b): Evolution of  $\gamma_{loc}$  as a function of time: — *Blue curves*: along the fingers (indicated by ticks in solid white lines in (a)); — *Red curves*: between two adjacent fingers, (as indicated by ticks in dotted-dashed white lines in (a)).  $b = 2.9$  mm.

purely elastic gel. This bulk instability is clearly different from the interfacial fingering observed in peeling [9, 10] or in some probe tak experiments [11]. For these instabilities, the wavelength increases linearly with the cell thickness, whereas here the inverse azimuthal wave number  $n_\theta^{-1} \sim b^{3/4}$  and the finger width  $\delta \sim b^{7/4}$ . These power laws are only indicative, in view of the limited range of values of  $b$  which can be explored, but such non-linear behaviors are in clear contrast with the interfacial crack case. As a matter of fact preliminary probe tak experiments suggest that the present instability is similar to the one reported by Shull et al. [15, 16]. Besides, these power laws also suggest the relevance of a second length scale in the problem, which should be the size  $D_0$  of the initial bubble. The  $1/D_0$  dependence of the critical strain is a strong indication in this direction.

Given the apparent versatility of the phenomenon, one may wonder why such elastic fingering instabilities have not been characterized in details before. One key point is that polyacrylamide adheres to the glass plates. Blowing or sucking air into the cell results in considerably deforming the layers of gel which are unable to glide. Fingering is a way for the system to release stresses except at the tip of the growing finger, and thus to minimize the elastic energy. Behind the finger tip, the layer of gel is still pulled hard, and will eventually break at a later stage. The two stages - instability and fracture - may be very close to one another if the material is not deformable enough. Fracture may even occur before the instability can be observed. Hence the conditions for observing the instability is to work with adhesive, highly deformable, still purely elastic materials, three conditions not so often met, when not looked for.

At the present stage, the precise mechanism of the instability remains unclear. However, the fact that the critical strain does not depend on the shear modulus suggests that there is no contribution other than elastic to the energy of the system. In particular, the interfacial tension between the injected fluid and the gel does not

play any role. We deal with a fully three dimensional elasticity problem, with interfaces deforming on large scales, which requires further investigation. Moreover, the hysteretic character of the phenomenon suggests that any linear stability analysis would be inefficient. A numerical non-linear approach similar to the one recently used to understand the formation of sulci in the Biot instability [22] may have to be developed. As a matter of fact, the instability considered in [22], which occurs when an incompressible elastomer is uniformly stressed laterally, is also purely elastic, and subcritical. The authors remark that when this instability is investigated without breaking translational symmetry at the surface, e.g. in a swollen, adhered layer of gel, then it naturally leads to extreme sensitivity, and subcritical instabilities connecting with buckling. And actually, the morphology of the patterns observed just before fracture are qualitatively reminiscent of buckling patterns described theoretically by Djondjorov and coworkers [23].

Finally, let us recall that fingering is commonly associated to liquids, while fracture is associated to solids. In this experiment, we show that fingering occurs also within the bulk of a purely elastic soft confined solid. Further experiments on different materials are currently being performed. In this context, it will be particularly interesting to analyze the crossover from our instability to a viscous fingering instability in a viscoelastic medium, as a function of the injected flow rate.

**Acknowledgements:** Nothing could have been done without the technical support of V. Padilla and C. Gasquet. This work was funded by the ANR (F2F project) and the Triangle de la Physique (FracHet project). Many thanks to F. Boulogne, for his kind hospitality at FAST/Orsay, and to R. Gy for providing plates made of Saint-Gobain float glass. We are also indebted to J.-P. Bouchaud, L. Mahadevan and S. Mora for enlightening discussions. E.B. also wishes to acknowledge the hospitality of CAS in Oslo, Norway.

---

\* Electronic address: baudouin.saintyves@cea.fr

- [1] P. Saffman and G. Taylor, Proc. of the Royal Soc. of London, Series A, Math. and Phys. Sci. **245**, 312 (1958).
- [2] D. Bensimon, L. P. Kadanoff, S. Liang, B. I. Shraiman, and C. Tang, Rev. of Mod. Phys. **58**, 977 (1986).
- [3] T. Hirata, Phys. Rev. E **57**, 1772 (1998).
- [4] S. S. Park and D. J. Durian, Phys. Rev. Lett. **72**, 3347 (1994).
- [5] E. Lemaire, P. Levitz, G. Daccord, and H. V. Damme, Phys. Rev. Lett. **67**, 2009 (1991).
- [6] P. Coussot, J. Fluid Mech. **380**, 363 (1999).
- [7] A. Lindner, P. Coussot, and D. Bonn, Phys. Rev. Lett. **85**, 314 (2000).
- [8] S. Mora and M. Manna, Phys. Rev. E **81**, 026305 (2010).
- [9] A. Ghatak, M. K. Chaudhury, V. Shenoy, and A. Sharma, Phys. Rev. Lett. **85**, 4329 (2000).
- [10] A. Ghatak, Phys. Rev. E **73**, 041601 (2006).

- [11] K. R. Shull, D. Ahn, W.-L. Chen, and C. M. Flani, *Macromol. Chem. Phys.* **199**, 489 (1998).
- [12] J. Nase, A. Lindner, and C. Creton, *Phys. Rev. Lett.* **101**, 074503 (2008).
- [13] M. Adda-Bedia and L. Mahadevan, *Proc. of the Royal Soc. of London, Series A, Math. and Phys. Sci.* **462**, 1 (2006).
- [14] T. Vilmin, F. Ziebert, and E. Raphael, *Langmuir* **26**, 32573260 (2010).
- [15] K. R. Shull, C. M. Flanigan, and A. J. Crosby, *Phys. Rev. Lett.* **84**, 3057 (2000).
- [16] R. E. Webber, K. R. Shull, A. Ross, and C. Creton, *Phys. Rev. E* **68**, 021805 (2003).
- [17] R. Nossal, *Rubber Chem. Tech.* **61**, 255 (1988).
- [18] B. A. Baker, R. L. Murff, and V. T. Milam, *Polymer* **51**, 2207 (2010).
- [19] L. Patterson, *J. Fluid Mech.* **113**, 513 (1981).
- [20] A. Ghatak and M. K. Cha, *The J. of Adh.* **83**, 679 (2007).
- [21] K. R. Shull and C. Creton, *J. of Pol. Sci. Part B: Pol. Phys.* **42**, 4023 (2004).
- [22] E. Hohfeld and L. Mahadevan, *Phys. Rev. Lett.* **106**, 105702 (2011).
- [23] P. Djondjorov, V. Vassilev, and I. Mladenov, *Int. J. of Mech. Sci.* **53**, 355 (2011).
- [24] Development of the instability in Setup 2. Acq rate 1500Hz, displaid at 15Hz  $V = 10\text{mm/s}$ ;  $D_0 = 2.2\text{cm}$ ;  $G = 180\text{Pa}$ . Note the elastic waves induced the burst of the fingers. .